

**REMARKS**

Reconsideration of the above-identified application in view of the foregoing amendments and following remarks is respectfully requested.

**I. Claim Status / Explanation of Amendments**

Claims 1, 3-4, 7, 15-19, and 22 are pending and were rejected, and claim 21 was previously withdrawn from consideration. By this paper, claim 1 is amended, and claim 22 is cancelled without prejudice or disclaimer. No new matter will be introduced into this application by entry of these amendments.

**II. Rejections Under 35 U.S.C. § 112:**

Claim 22 was rejected pursuant to 35 U.S.C. § 112, first paragraph, as allegedly failing to comply with the written description requirement. [6/7/2010 Office Action, p. 2]. In order to expedite prosecution of this application, claim 22 has been cancelled without prejudice or disclaimer. Accordingly, the Section 112 rejection of claim 22 has been rendered moot.

**III. Rejections Under 35 U.S.C. § 103:**

Claims 1, 3-4, 7 and 15-19 were rejected under 35 U.S.C. § 103(a) as allegedly being unpatentable over U.S. Patent No. 4,742,110 to Sakashita, et al. (hereinafter "Sakashita"). [6/7/2010 Office Action, p. 3]. Applicants respectfully traverse the rejection since, as set forth in detail below, Sakashita fails to teach or disclose a coating composition consisting of a polyimide or polyamide-imide binder resin. That is, "polyamide or polyamide-imide" resin recited in claim 1 is patentably different from "the polyamide composition" described in Sakashita. Indeed, "the polyamide composition" described in Sakashita is inadequate for use as a sliding film for a sliding part.

It is well known that the temperature of a sliding part rises due to friction forces generated by its sliding actions. By way of example, for the sliding parts in a compressor, temperatures ranging from 150°C to 180°C are generally anticipated, and even 200°C can be obtained under more onerous conditions. Thus, a sliding film is designed to endure such high temperatures.

The "Comprehensive Composite Materials" handbook discloses glass transition temperatures of polyimide, polyamideimide, and polyamide<sup>1</sup>. "[W]hen the glass transition is exceeded, brisk molecular movement takes place, resulting in remarkable decrease in elastic modules and conspicuous volume expansion. Thus, the melting point with crystalline polymers and the glass transition temperature with amorphous polymers become criteria to determine heat resistance."<sup>2</sup> Since the glass transition temperature of polyamide is 70°C, polyamide is not suitable for use in the sliding film for the sliding part at all.

Indeed, the Sakashita patent itself shows the polyamide composition is not suitable for use in the sliding film for a sliding part. Directing the Examiner's attention to Examples 1-7 described in Table 4 of Sakashita patent, the distortion temperature for Example 5 is 173°C, but the distortion temperatures for Examples 1-4 and 6 are on the order of 140°C which is fairly lower than Example 5. A composition which distorts at 140°C or 173°C is not adapted to be used as a sliding film for the sliding part. Example 7 is exceptional in that the distortion temperature is indicated as being greater than 250°C. However,

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<sup>1</sup> A. Kelly and C. Zweben, "Comprehensive Composite Materials," Elsevier Science, (2000) at J. D. Muzzy, Thermoplastics - Properties, Table 1, Part B. See Exhibit A hereto.

<sup>2</sup> Booklet on Engineering Plastics, The Japan Engineering Plastics Association (1991) at p. 14. See Exhibit B hereto.

Example 7 included 20% of glass fibers therein so that the distortion temperature of Example 7 was improved. Thus, Example 7 does not show that a polyamide composition has a high distortion temperature.

The "deflection temperature under load" is used as a measure of heat resistance, and refers to the temperature at which resins are brought to deformation. With fiber reinforced polymers especially crystalline polymers, the molecular movement at the glass transition temperature is restricted, exhibiting a behavior that the materials apparently withstand as applied load. For this reason, apart from the deflection temperature under load, long-term continuous service temperatures are determined.<sup>3</sup>

Referring to Figure 2.2.1 of the Booklet on Engineering Plastics (see Exh. B at p. 15), the figure shows a relation between a continuous usage temperature and a deflection temperature under load. Figure 2.2.1 discloses that the continuous usage temperatures for polyimide and polyamide-imide are greater than 230°C, but the continuous usage temperatures for nylon and nylon-GF (glass-fiber-enforced nylon) is lower than 120°C. From this figure, it is clear that nylon cannot be used under a high temperature.

Sakashita patent describes that imide-based resins such as polyimides and polyamide-imides may be incorporated into the polyamide composition. See column 6, lines 19-22. However, the description is that these imide based resins are merely exemplified as "stabilizers, plasticizers, parting agents, lubricants and fillers." See column 5, line 68 of column 6, line 1. A person of ordinary skill in the art would believe that the addition of the imide-based resin is to improve the

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<sup>3</sup> Booklet on Engineering Plastics, The Japan Engineering Plastics Association (1991) at pp. 14-15. See Exhibit B hereto.

properties of the composition by adding solid filler which does not melt under high temperature. Prior art patents disclose that polyimide fibers are added as a filler to improve fire resistance or flame retardant property.<sup>4</sup>

The imide-based resin of the present invention also acts as a binder resin, when it is dissolved, which securely adheres the sliding film to the sliding part body, or solid lubricants and/or titanium oxide powders contained in the coating composition to the sliding film. On the other hand, even if it is suggested in Sakashita patent that imide-based resins are added, the imide-based resins does not function as a binder resin.

Accordingly, Sakashita fails to teach, disclose, or suggest a "wear resistant and seizure resistant film wherein the film is formed of a coating composition comprising a binder resin, which is polyimide or polyamide-imide" as recited in Applicants' amended claim 1. Applicants submit that claim 1 is patentable over Sakashita for at least this reason. Claims 3-4, 7, and 15-19 are also deemed to be in condition for allowance by way of their direct or indirect dependence on claim 1. Applicants therefore respectfully request that the Section 103 rejection of claims 1, 3-4, 7, and 15-19 be withdrawn and submit that these claims are now allowable for the above reasons. Early, favorable action in that regard is respectfully requested.

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<sup>4</sup> See Kaide U.S. Patent No. 4,255,318 at column 4, lines 39 and 40 [Exh. C hereto]; Inagaki U.S. Patent No. 4,191,675 at column 2, lines 22-27 [Exh. D hereto]; and Mitani U.S. Patent No. 3,937,689 at column 6, lines 10-16 [Exh. E hereto].

**CONCLUSION**

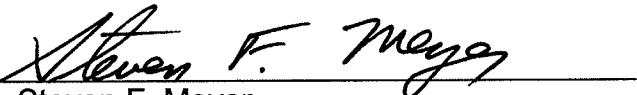
In light of the foregoing claim amendments and response, Applicants respectfully submit that all of the pending claims are now in condition for allowance. Thus, it is respectfully requested that the rejections under §§ 112 and 103 be withdrawn, and all the claims be allowed such that this application may expeditiously pass to issue.

**AUTHORIZATION**

The Commissioner is hereby authorized to charge the fee for a one-month extension of time or credit any overpayment for an extension of time to Deposit Account No. **504827**, Order No. **1004378.51670**. The Commissioner is hereby authorized to charge any additional fees which may be required for consideration of this Document to Deposit Account No. **504827**, Order No. **1004378.51670**.

Respectfully submitted,  
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Dated: October 7, 2010

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# **EXHIBIT A**

## 2.3

### Thermoplastics – Properties

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- 2.3.1 INTRODUCTION
- 2.3.2 GENERAL FEATURES
- 2.3.3 GUIDELINES FOR SELECTION OF POLYMERS AND PROPERTIES
- 2.3.4 THERMOPLASTIC GROUPS
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  - 2.3.4.1.2 *Styrenics*
  - 2.3.4.1.3 *Vinyls*
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  - 2.3.4.1.9 *Polyethers*
  - 2.3.4.1.10 *Sulfur Containing polymers*
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- 2.3.5 ACKNOWLEDGEMENTS
- 2.3.6 REFERENCES

#### 2.3.1 INTRODUCTION

The primary objective of this chapter is to present representative properties of thermoplastics. First, some general comparisons with thermosetting matrices are presented since most high performance composites have thermosetting matrices. Next, eleven groups are established for dividing the presentation of 40 different types of thermoplastics. Then the properties of these thermoplastic types are presented.

Not all types of thermoplastics are included in this chapter. Thermoplastic rubbers have been excluded since these rubbers have low elastic moduli. Usually the reason for reinforcing a thermoplastic is to increase its stiffness and strength. From this perspective a thermoplastic rubber represents a low starting point. If the property desired is high impact strength or durability, then reinforcing a thermoplastic rubber should be considered.

#### 2.3.2 GENERAL FEATURES

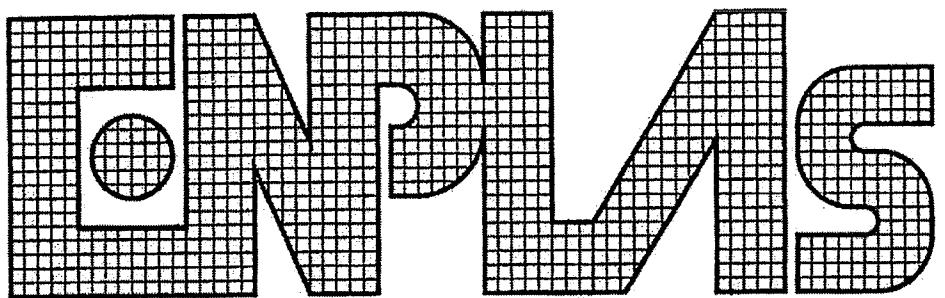
The mechanical properties of polymers are sensitive to temperature changes. Figure 1 illustrates the change in modulus with respect to temperature for an amorphous thermoplastic. Below its glass transition temperature ( $T_g$ ) the modulus is relatively constant with a value close to 2.8 GPa (0.4 ksi). As the temperature increases above  $T_g$  the modulus drops roughly three orders of magnitude to 0.28 GPa (0.4 ksi) as the polymer becomes rubbery. If the molecular weight of the

Table 1, Part B  
Thermoplastic Selling Prices and Transition Temperatures

Section	Type	Abbrev.	Transition			
			Prices			Temperatures
			\$/lb	\$/kg	Tg (°C)	Tm (°C)
2.3.4.6	Polyesters, continued					
	20 Polyarylate	PAR	2.40	5.29	190	am*
	21 Liquid Crystal Polyester	LCP	12.00	26.45		400
2.3.4.7	Polyamides (Nylons)					
	22 Nylon Type 6	N6	1.30	2.87	60	220
	23 Nylon Type 66	N66	1.40	3.07	70	260
	24 Nylon Type 11	N11	3.35	7.38	-70	185
	25 Nylon Type 12	N12	3.30	7.26		190
	26 Polyphthalamide	PPA				310
	27 Polyamideimide	PAI	26.10	57.52	275	am*
2.3.4.8	Polyimides					
	28 Polyetherimide	PEI	6.41	14.13	215	am*
	29 Polyimide	PI			320	385
2.3.4.9	Polyethers					
	30 Polyacetal	POM	1.25	2.76	-75	175
	31 Polycarbonate	PC	1.55	3.42	150	am*
	32 Polyphenylene oxide blend	PPO	1.80	3.97	110-190	am*
	33 Polyaryletherketone	PAEK	29.50	65.02	138	320-380
	34 Polyetheretherketone	PEEK	33.00	72.73	145	335
2.3.4.10	Sulfur Containing Polymers					
	35 Polyphenylene sulfide	PPS			90	285
	36 Polysulfone	PSF	4.40	9.70	190	am*
	37 Polyethersulfone	PES	4.40	9.70	225	am*
	38 Polyarylsulfone	PAS	4.40	9.70	220	am*
2.3.4.11	Additional Thermoplastics					
	39 Acrylonitrile copolymer	ANC	1.25	2.76	95	am*
	40 Polyurethane	TPU	2.10	4.63		am*

am\* = amorphous

# **EXHIBIT B**



**Booklet  
on  
Engineering  
Plastics**

The Japan Engineering Plastics Association

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ments calling for higher heat resistance of plastics, new high heat resistant polymers have been brought into existence one by one.

Polymers having higher heat resistance should have greater bonding energy with main chains, one of necessary conditions. That is, polymers having phenylene groups and with rigid structure can be pointed out to have higher heat resistance.

More in detail, these heat resistant polymers have common features as follows:

- 1) Polycondensation materials are mainstream.
- 2) In the backbone of main chains, the materials have *p*-phenylene groups.

#### (1) Melting point ( $T_m$ ) and Glass Transition Temperature ( $T_g$ )

Polymers having a stereoregular structure are readily crystallized, and become crystalline polymers with a melting point. The melting point ( $T_m$ ) has a following relation between melt enthalpy ( $\Delta H$ ) and melt entropy ( $\Delta S$ ):

$$T_m = \Delta H / \Delta S$$

Where,  $\Delta H$  is an intermolecular force, and for example, when there are hydrogen bonds, it becomes greater.  $\Delta S$  becomes smaller when main chains have symmetrical structure, are crosslinked, and have higher rigidity.

On the contrary, the polymers such as having twisted phenylene groups become amorphous to have a glass transition temperature ( $T_g$ ).

Generally, below the glass transition temperature, the molecular movement is frozen with less change in properties, but when the glass transition temperature is exceeded, brisk molecular movement takes place, resulting in remarkable decrease in elastic modulus and conspicuous volume expansion.

Thus, the melting point with crystalline polymers and the glass transition temperature with amorphous polymers become criteria to determine heat resistance.

## 2.2.2 Practical Heat Resistance of Polymers

### (1) Short-Term Heat Resistance

The melting point and glass transition temperature of polymers are the points of behavior accompanied with phase changes inherent in polymers, and it is clear these are not the criteria by which polymers can be understood to be capable of use practically.

What's realistically used as a measure of heat resistance is the deflection temperature under load (DTUL, formerly called HDT).

The determination is comprised of, heating a testing specimen under a load of 1.8 or 0.5 MPa, the temperature at which the specimen starts to deform. Polymers themselves deform at the temperature, so that it is natural that the temperature range in which the polymers can be used is less than the stress level. Normally, in case of short-term usage, it appears that the temperature about 10°C less than the level is considered the upper-limit usable temperature of polymers.

### (2) Long-Term Heat Resistance

Deflection temperature under load refers to the temperature at which resins are brought to deformation, implying that the temperature cannot be said to be practical. Especially, in such a case when it is intended to use resins for a long period, it is necessary to clarify the heat resistant properties resins possess. With fiber reinforced polymers, especially crystalline polymers, the molecular movement at the glass transition temperature is restricted, exhibiting a behavior that the materials apparently withstand a load applied. For the reason, apart from the deflection temperature under load, long-term continuous service temperatures are determined.

As for the long-term continuous service temperature, a method of determination is decided by UL specifications as a criterion, defining that the service temperature refers to the temperature at which :

material's physical properties are reduced by half through exposure to elevated temperatures for several ten thousands hours. With focus placed on the usage results with a variety of polymers, the long-term continuous service temperatures are obtained, the relation of which with deflection temperatures under load is shown in Fig. 2.2.1.

What should be taken into account with the figure is that while in case of crystalline polymers, heat deflection temperature under load increases remarkably by reinforcing with glass fiber, the long-term continuous serviceable temperatures are similar to the level with unreinforced grades. Special attention should be paid to such resins as polyamide, PBT, etc.

On the other hand, PTFE, PEEK, and some others have heat deflection temperature under load not so high with unreinforced grades, but when used under a smaller load, have heat resistance up to 250-260°C.

The temperature for long-term continuous uses at which polymer materials can withstand differs depending on the conditions of how the load is applied, how the materials are used, and on environment and heat dissipation conditions, and other factors, so that most suitable resins should be selected with sufficient considerations placed on the level of requirements on specifications.

In case of amorphous polymers, additionally, the deflection temperature under load increases only slightly with the addition of glass fiber.

### (3) Soldering Heat Resistance

Packaging of electric/electronics parts applied to printed circuit boards was mainly carried out formerly by directly immersing these parts into a molten soldering bath, but the method today has been shifted to the reflow soldering method.

Generally, with the former case, the soldering bath has a temperature of 260°C, indicating that without having the heat resistance exceeding the

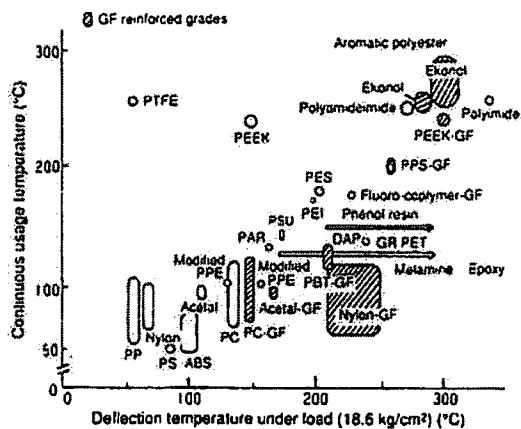


Fig. 2.2.1 Correlation between heat deflection temperature under load, and temperature

level, terminals of soldering parts become loosened, or resin materials are molten during the soldering process, an inconvenient phenomenon.

The guidelines of heat resistance are heat deflection temperature being 260°C as feasible, because of the extremely short time for the soldering process. Some large sized parts have greater heat capacity, and before the temperature reaches 260°C, the soldering process could be finished, so that it appears materials having a heat deflection temperature under load in a range of 220-230°C could withstand.

On the other hand, however, in order to shorten treating time, to secure the soldering process, and not to leave excess molten solder on printed circuits, there is a tendency to apply soldering at still higher temperature. As a result, even with the reflow method, resin materials without the heat deflection temperature under load above 260°C have come to be incapable of withstanding.

Reflow soldering is a method brought to practice in accordance with miniaturization and mass production of electric/electronics parts and components, and is often used for the production of chip parts. These parts are treated under circumstances heated

# **EXHIBIT C**

## United States Patent [19]

Kaide et al.

[11] 4,255,318

[45] Mar. 10, 1981

## [54] FIRE-RETARDANT PUTTY-LIKE COMPOSITIONS

[75] Inventors: Tamotsu Kaide; Toshikazu Gozen, both of Takarazuka; Jimichi Taniguchi, Takatsuki; Yutaka Ohta, Amagasaki, all of Japan

[73] Assignee: Dainichi-Nippon Cables, Ltd., Amagasaki, Japan

[21] Appl. No.: 58,334

[22] Filed: Jul. 17, 1979

[51] Int. Cl. 7/14 C08K

[52] U.S. Cl. 260/42.18; 260/42.34; 260/DIG. 24

[58] Field of Search 260/42.18, 42.34, 23.7 H, 260/31.8 DR, 30.6 R, DIG. 24

## [56] References Cited

## U.S. PATENT DOCUMENTS

3,843,592 10/1974 Perkins ..... 260/42.34  
4,138,537 2/1979 Dembowski et al. ..... 260/42.34

Primary Examiner—Lewis T. Jacobs

*Attorney, Agent, or Firm*—Pollock, Vande Sande & Priddy

## [57] ABSTRACT

A composition for filling the spaces in bores penetrating walls and floors of buildings and having electric wires and cables extending therethrough or for filling the clearances at the joints of interior finishing materials of buildings. The composition comprises (a) 100 parts by weight of a curable polychloroprene in a liquid state at room temperature, (b) about 200 to about 700 parts by weight of a hydrated metallic oxide, and (c) about 20 to about 100 parts by weight of a heat-resistant fibrous material. The components (b) and (c) are contained in a combined amount of at least about 250 parts by weight per 100 parts by weight of the component (a). The composition will not soften, sag or drip in molten drops even when subjected to the high-temperature conditions of a fire, and gives a tough residual product retaining the original shape when burned and ashed, effectively preventing the spread of fire and assuring outstanding smoketightness.

25 Claims, No Drawings

4,255,318

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curing agent selected from the above are used according to the end group of polychloroprene.

These curing agents are used in an amount of about 0.5 to about 20 parts by weight per 100 parts by weight of the liquid polychloroprene.

Preferable among the liquid polychloroprenes exemplified above are those having a viscosity of about 5,000 to about 500,000 cps, especially about 10,000 to about 300,000 cps, at room temperature (25° C.). With respect to the end group, preferable are those having an alkylxanthate group in which the alkyl has 2 to about 10 carbon atoms such as ethyl, propyl, butyl etc. It is also desirable to use those having a hydroxyl end group conjointly with a diisocyanate such as tolylenediisocyanate serving as a curing agent. It is more desirable to use a liquid polychloroprene having at least one kind of the alkylxanthate end groups as admixed with a liquid polychloroprene having at least one hydroxyl end group in an amount of up to about 100 parts by weight per 100 parts by weight of the former.

According to this invention, the liquid polychloroprenes given above are usable, with or without any of the curing agents mentioned above when so desired. Preferable polychloroprenes are those satisfying curing properties as determined by the following test method.

Test method: 100 parts by weight of the component (a) is admixed with 400 parts by weight of  $\text{Al}_2\text{O}_3 \cdot 3\text{H}_2\text{O}$  (mean particle size: 3.5  $\mu\text{m}$ ) and 30 parts by weight of glass fiber (mean diameter: 13  $\mu\text{m}$ , mean length: 6 mm) and the mixture is kneaded into a putty-like composition, which is then heated at 250° C. for 30 minutes. The component (a) is acceptable when the composition, after the heating, is up to about 1, preferably up to about 0.5 in cone penetration value evaluated in accordance with JIS A 5752-1966 (mm/150 g, 5 sec, at 20° C.).

The hydrated metallic oxide (or hydroxide of metal) serving as the component (b) of the present compositions is in the form of a fire-retardant or nonflammable powder having a mean particle size of up to about 100  $\mu\text{m}$ , preferably up to about 80  $\mu\text{m}$ , and having a heat loss of at least about 8% by weight, preferably, at least about 20% by weight obtained by the following formula:

$$\text{heat loss (O/O)} = A - B/4 \times 100$$

where

A: initial weight of sample

B: constant weight of sample after heating at a temperature of  $400 \pm 20^\circ \text{C}$ .

Examples of useful hydrated metallic oxides are hydrated aluminas, represented by the formula  $\text{Al}_2\text{O}_3 \cdot n\text{H}_2\text{O}$  (n being 0.5-about 6), such as  $\text{Al}_2\text{O}_3 \cdot \frac{1}{2}\text{H}_2\text{O}$ ,  $\text{Al}_2\text{O}_3 \cdot \text{H}_2\text{O}$ ,  $\text{Al}_2\text{O}_3 \cdot 2\text{H}_2\text{O}$ ,  $\text{Al}_2\text{O}_3 \cdot 3\text{H}_2\text{O} \cdot \text{Al}(\text{OH})_3$ , etc. and hydrated magnesias such as  $\text{Mg}(\text{OH})_2$ , etc.

It is preferably to use a hydrated metallic oxide comprising at least two portions which differ in particle size, or to conjointly use at least two kinds of hydrated metallic oxides which differ in particle size. Stated more specifically, the component (b) comprises at least two portions one of which has a mean particle size of about 10 to about 100  $\mu\text{m}$ , preferably about 10 to about 80  $\mu\text{m}$ , the other portion being up to about 10  $\mu\text{m}$  in mean particle size.

The hydrated metallic oxides serving as the component (b) are used in an amount of about 200 to about 700 parts by weight, preferably about 250 to about 450 parts by weight, per 100 parts by weight of the component (a). With less than about 200 parts by weight of the

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component (b) present, the putty-like composition has greatly increased flowability, is prone to deformation when applied even at room temperature, is liable to soften and drop when subjected to the heat of fires and gives a brittle residue when ashed. With use of more than about 700 parts by weight of the component (b), the ingredients (a), (b) and (c) will have reduced compatibility when they are mixed, while the resulting composition is not satisfactorily applicable to spaces, bores or the like and affords low airtightness even at room temperature if filled in place.

When the component (b) comprises at least two portions of different particle sizes, the portion up to about 10  $\mu\text{m}$  in mean particle size is used in an amount of about 10 to about 500 parts by weight per 100 parts by weight of the other portion with a mean particle size of 10 to about 100  $\mu\text{m}$ .

According to this invention it is advantageous to use the component (b) in combination with particles, smaller than about 10  $\mu\text{m}$ , of at least one of clay, zinc borate, bentonite, talc, diatomaceous earth, calcium carbonate and mica in an amount of up to about 80% by weight, preferably up to about 50% by weight, based on the component (b). The composition will then afford a residual product of enhanced toughness when ashed. Among the above-mentioned materials, clay, zinc borate and bentonite are especially advantageous to use.

The heat-resistant fibrous materials useful as the component (c) of the present compositions are inorganic fibers, and organic polymeric fibers which will not thermally deform at temperatures of lower than about 250° C. Such fibers are up to about 100  $\mu\text{m}$ , preferably about 0.5 to about 50  $\mu\text{m}$ , in mean diameter and up to about 30 mm, preferably about 1 to about 20 mm, in mean length.

Examples of useful inorganic fibers are glass fiber, asbestos fiber, carbon fiber, etc. Examples of suitable organic polymeric fibers are phenolic resin fibers, polyimide fiber, polyamide-imide fiber, etc. Among these fibers, glass fiber and asbestos fiber are preferable. A mixture of glass fiber and asbestos fiber is more preferable.

The heat-resistant fibers serving as the component (c) are used in an amount of about 20 to about 100 parts by weight, preferably about 20 to about 60 parts by weight, per 100 parts by weight of the component (a). With less than about 20 parts by weight of the component (c), the putty-like composition, when exposed to the high temperature of a fire, is liable to soften and sag, and also fails to give a tough ashed product. If used in an amount of more than about 100 parts by weight, the component (c) will be less compatible with the other ingredients when formulated into a putty-like composition, while the composition is not satisfactorily applicable and provides impaired airtightness at room temperature if used.

When the heat-resistant fibrous material (c) comprises a mixture of glass fiber and asbestos fiber, it is suitable to use about 10 to about 300 parts by weight of asbestos fiber per 100 parts by weight of glass fiber.

As described above, the fire-retardant putty-like compositions of the present invention consist essentially of 100 parts by weight of the component (a), about 200 to about 700 parts by weight of the component (b) and about 20 to about 100 parts by weight of the component (c). It is also critical that the compositions contain the component (b) and the component (c) in a combined amount of at least about 250 parts by weight per 100

# **EXHIBIT D**

## United States Patent [19]

Inagaki et al.

[11] 4,191,675  
[45] Mar. 4, 1980[54] FIRE SPREADING INHIBITOR  
COMPOSITION[75] Inventors: Yutaka Inagaki; Hironaga  
Matsubara; Kojiro Ishise, all of  
Osaka, Japan[73] Assignee: Sumitomo Electric Industries, Ltd.,  
Osaka, Japan

[21] Appl. No.: 778,281

[22] Filed: Mar. 16, 1977

## [30] Foreign Application Priority Data

Mar. 16, 1976 [JP] Japan ..... 51/28879

[51] Int. Cl.<sup>2</sup> ..... C09K 3/28  
[52] U.S. Cl. ..... 260/29.3, 252/8.1;  
260/29.6 NR; 260/38; 260/DIG. 24[58] Field of Search ..... 260/29.3, 29.6 NR, DIG. 24;  
252/8.1[56] References Cited  
U.S. PATENT DOCUMENTS2,536,978 1/1951 Fordemwalt ..... 260/15  
3,630,764 12/1971 Shannon ..... 106/15  
3,642,531 2/1972 Peterson ..... 428/921  
3,928,210 12/1975 Peterson ..... 260/29.6 R XPrimary Examiner—Herbert B. Guynn  
Attorney, Agent, or Firm—Sughrue, Rothwell, Mion,  
Zinn and Macpeak

## [57] ABSTRACT

A fire spreading inhibitor composition comprising about 20 to about 40% by weight of an emulsion of a synthetic resin, about 1 to about 15% by weight of non-fusible organic fibers, about 20 to about 70% by weight of an inorganic powder at least about 50% of which is clay and/or zinc borate, and about 5 to 20% by weight of a halogenated hydrocarbon, all percentages being based on the dry weight as a coating.

7 Claims, 2 Drawing Figures

4,191,675

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## FIRE SPREADING INHIBITOR COMPOSITION

## BACKGROUND OF THE INVENTION

## 1. Field of the Invention

This invention relates to a fire spreading inhibitor composition.

## 2. Description of the Prior Art

Building materials and cables, etc. made of combustible materials, especially combustible plastics such as polyethylene or polyvinyl chloride, readily burn in the event of fire, and the fire may spread to other objects and cause the danger of conflagration of houses and other facilities. It has been the previous practice, therefore, to coat fire spreading inhibitor compositions (often referred to "fire protective composition" or "fire retardant" by one skilled in the art) on the surfaces of such combustible materials to prevent the spreading of fire.

## SUMMARY OF THE INVENTION

An object of this invention is to provide a fire spreading inhibitor composition with a superior performance not heretofore attainable.

The present invention provides a fire spreading inhibitor composition comprising about 20 to about 40% by weight of an emulsion of a synthetic resin, about 1 to about 15% by weight of non-fusible organic fibers, about 20 to about 70% by weight of an inorganic powder at least 50% of which is clay and/or zinc borate, and about 5 to about 20% by weight of a halogenated hydrocarbon, all percentages being based on the dry weight of the composition as a coating.

## BRIEF DESCRIPTION OF THE DRAWINGS

FIGS. 1 and 2 show equipment arrangements used in performance of evaluations conducted in the examples.

## DETAILED DESCRIPTION OF THE INVENTION

The fire spreading inhibitor composition of this invention is especially effective against objects requiring flexibility, weatherability and water resistance, such as cables. For simplicity, features and advantages of the invention will be described hereinbelow with special regard to cables. Needless to say, however, the composition of this invention is also effective against other combustible objects such as building materials in general.

Polyethylene and polyvinyl chloride, for example, have been used heretofore as coverings or jackets for insulators and sheaths, etc. of electric wires and cables. In the event of fire, these combustible coverings will burn and permit the propagation of the flame and are highly likely to cause damage to a control room and other electric facilities, and thus increase the loss due to fire. In order to prevent such a situation, attempts have been made to coat electric wires and cables with foamable or non-foamable fire spreading inhibitor compositions. For example, U.S. Pat. No. 3,642,531 discloses a typical example of a conventional fire spreading inhibitor composition which comprises a synthetic resin emulsion, inorganic fibers such as asbestos, and a halogenated hydrocarbon as main ingredients and an inorganic filler such as calcium carbonate or silica as an additional ingredient.

However, the inorganic fibers used in such an inhibitor composition may cause operational troubles during the manufacture of the inhibitor composition. Usually,

glass and asbestos are used as the inorganic fibers. Both of these fibers tend to adversely affect working personnel during manufacture, and therefore not preferred. In particular, asbestos is detrimental to the health of working personnel, and the spraying of a composition containing asbestos is lawfully prohibited under some conditions to eliminate hazardous working conditions.

Furthermore, the fire spreading inhibition effect of such a conventional composition is not entirely satisfactory. When a flame comes into contact with a cable coated with the inhibitor composition, cracks occur in that portion of the cable which is vertically oriented whereby the combustible covering, such as polyethylene, inside the coating comes out and catches fire.

All of these defects have been successfully eliminated with the fire spreading inhibitor composition of the invention.

A first feature of the invention lies in the use of organic fibers which are non-fusible, i.e., do not melt when heated. Examples of suitable organic fibers are phenolic resin fibers such as "Kaynol" (registered trademark for a product of Japan Kaynol Co., Ltd.), polyimide fibers, polyamideimide fibers, and completely aromatic polyamide fibers such as "Nomex" (tradename, a product of E. I. du Pont de Nemours & Co.). Phenolic resin fibers are especially preferred. It has been found that these organic fibers used in place of the inorganic fibers fully meet the characteristics required of fireproofing composition, such as strength, and exhibit a satisfactory fire spreading inhibition effect, and moreover, the problem of hazardous working conditions can be solved.

When the amount of the organic fibers is less than about 1% by weight, cracks may occur in the coated composition during a fire. If the amount is more than about 15% by weight, the coatability of the composition becomes poor. For operational purposes, amounts of about 1 to 5% by weight are preferred.

A second characteristic feature of the present invention is the use of an inorganic powder at least about 50% of which is clay and/or zinc borate. When an ordinary inorganic powder such as calcium carbonate or silica is used as disclosed in Japanese Patent Application (OPI) No. 4979/73, a cracking of the coating of the composition formed on a vertically placed cable occurs upon contact with a flame, and combustible materials such as polyethylene inside the cable comes out and catch fire. In such a case, the coating of the fire spreading inhibitor composition becomes useless since the combustible material is exposed and is no longer covered. After extensive investigations about this point, it was found that when a powder comprising clay and/or zinc borate is used, the heat of the flame causes the clay and/or zinc borate to form a hard shell upon contact with the flame, and a flowing of the combustible materials such as polyethylene to the outside is prevented. It was also found that if the amount of the powder is about 20 to about 70% by weight, and at least about 50% of the entire powder is clay and/or zinc borate, even the combined use therewith of another inorganic powder such as aluminum hydroxide or calcium carbonate can lead to the formation of a sufficiently hard shell and prevent the fire from spreading.

When aluminum hydroxide is used in combination as an inorganic powder, the amount of smoke occurring during burning is reduced, and the fire spreading inhibi-

# **EXHIBIT E**

## United States Patent [19]

Mitani et al.

[11] 3,937,689

[45] Feb. 10, 1976

## [54] RADICAL INITIATOR-CURABLE DIALYL ESTER COMPOSITIONS AND CURED RESINS OBTAINED THEREFROM

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[22] Filed: July 1, 1974

[21] Appl. No.: 484,871

[52] U.S. Cl. 260/78.5 UA; 260/78.4 UA; 260/875  
[51] Int. Cl. 2 C08F 218/16; C08F 218/18;

C08F 222/26

[58] Field of Search ..... 260/78.4 UA, 78.5 UA

## [56] References Cited

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3,113,123	12/1963	Heiberger et al. ....	260/78.5 UA
3,424,729	1/1969	Lanaka et al. ....	260/78.4 UA
3,455,888	7/1969	Thomas.....	260/78.5 UA

Primary Examiner—John Kight, III

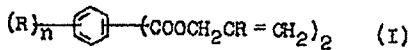
Attorney, Agent, or Firm—Sherman &amp; Shalloway

## [57] ABSTRACT

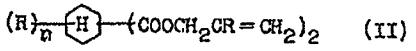
A radical initiator-curable dialyl ester composition consisting essentially of 80 – 100% by weight of the following components (A) and (B):

A. a prepolymer wherein

1. 70 – 100 mole % of its structural units are derived from a dialyl ester selected from the group consisting of  
a. a dialyl ester of benzenedicarboxylic acid having the formula (I)

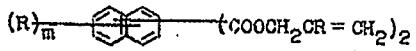


b. a dialyl ester of cyclohexanedicarboxylic acid having the formula (II)



c. a mixture of the above (a) and (b); and  
2. 30 – 0 mole % of its structural units are derived from other compounds selected from monomeric dialyl esters of dibasic acids and other monomers and polymers, said compound being copolymerizable with said (a) – (c) under the conditions of forming said prepolymer; and

B. a monomer of a dialyl ester of naphthalenedicarboxylic acid having the formula (III)



(III)

3 Claims, No Drawings

3,937,689

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position results to make its comminution impossible, or troubles such as form retention difficulties occur due to the blocking phenomenon during the composition's storage. On the other hand, when the invention monomer (B) is used, these troubles at the time of comminution or storage do not happen at all, and the improvement in the flow is readily attained.

Further, the invention composition can also contain minor amounts of other monomers or polymers (C) that are copolymerizable with said prepolymer and/or monomer at the conditions under which said composition is cured.

As specific examples of these monomers and polymers (C), mention can be made of the vinyl monomers such as acrylonitrile, styrene and methacrylate; the dialyl and dimethallyl esters of such dibasic acids as orthophthalic acid, isophthalic acid, terephthalic acid, methylorthophthalic acid, methylisophthalic acid, methylterephthalic acid, trans- and cis-hexahydroorthophthalic acids, trans- and cis-hexahydroisophthalic acids, and trans- and cis-hexahydroterephthalic acids; the dialyl and dimethallyl esters of such dibasic acids as diphenylmethanedicarboxylic acid, diphenyldicarboxylic acid, diphenylsulfonedicarboxylic acid, hexachloroendomethylenetetrahydrophthalic acid, maleic acid, fumaric acid, sebacic acid and adipic acid; the prepolymers of these dibasic acid dialyl esters and/or dimethallyl esters; the prepolymers of the naphthalenedicarboxylic acid dialyl esters; the monoallyl esters of monobasic acids, such as allyl benzoate, allyl methacrylate and allyl naphthoate; the polyallyl esters such as trimellitic acid triallyl ester, triallyl cyanurate, triallyl disocyanurate, triallyl phosphate and naphthalenetetracarboxylic acid tetraallyl ester; and 1,2- and 1,4-polybutadiene and the unsaturated polyesters.

These monomers and/or polymers (C) can be used in an amount of 20 - 0% by weight, preferably 10 - 0% by weight, of the total composition. Of these monomers, in the case of especially dialyl orthophthalate, dialyl isophthalate and dialyl terephthalate, which are monomers that are liquid at room temperature, it is best that these be used in an amount of not more than 10% by weight, since adverse effects will be had on the resulting resin when used in excess.

The invention composition is curable with the radical initiators. While the amount used of the radical initiator and the curing temperature are suitably varied in accordance with the makeup of the composition and the class of initiator used, the amount usually used is preferably about 0.5 - 5% by weight based on the total of the aforesaid components (A) and (B) and, as the case may be, the component (C). And a curing temperature usually in the range of about 100 - 200°C. is used. As this radical initiator, preferably used are such organic peroxides as di-tertiary-butylperoxide, ditertiary-allylperoxide, benzoyl peroxide, tertiary-butylperbenzoate, ditertiary-butylhydroperoxide and dicumylperoxide. Irradiation with electron beam is also effective. For instance, when curing the invention composition on the surface of a substrate, the polymerization and cure of the composition can be accomplished in an extremely short period of time by irradiation with electron beam of the order of 0.5 - 70 Mrad, thus forming a cured coating on the surface of the substrate.

Further, the properties of the resulting cured resin can be improved still further by incorporating in the resinous composition of the invention, as required, such additives as fillers, mold releasing agents, poly-

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merization accelerators, polymerization retarders, stabilizers, pigments and silane coupling agents to an extent that does not impair the effects of the invention composition.

As the foregoing fillers, included are the inorganic fillers such, for example, as mica, asbestos, powdered glass, silica, clay, titanium dioxide, magnesium oxide, alumina, asbestos fibers, silica fibers, glass fibers, silicate glass fibers, alumina fibers, carbon fibers, boron fibers, baryllium fibers, steel fibers and whiskers; and the organic fillers such as polyethylene, polypropylene, polyvinyl chloride, polyvinyl fluoride, polymethyl methacrylate, aliphatic and aromatic polyamides, polyimides, polyesterimides, polyamideimides, pulp, acrylic fibers, polyester fibers, such as polyethylene terephthalate, cotton and rayon. The amounts in which these fillers are used can be varied over broad limits, but preferred is an amount ranging from 20 to 500% by weight based on the total of the prepolymer (A) and monomer (B) and, as the case may be, the copolymerizable other monomer and/or polymer (C).

As the properties of the filler have an effect on the properties of the cured product, the class and amount of the filler used are chosen in accordance with the properties desired in the intended cured product. For instance, when the requirement is that the cured product be especially resistant to heat, an inorganic filler or an organic filler excelling in thermal resistance (e.g. polyvinyl fluoride, polybutadiene, an aromatic polyamide, polyimide, polyamideimide and polyesterimide) is chosen. When using these fillers, their mixing with the prepolymer can be accomplished readily and uniformly by a procedure consisting of, say, dissolving the foregoing prepolymer in an organic solvent.

As the solvent for use in this case, included are such, for example, as the aromatic hydrocarbons as benzene and toluene, the ketones as acetone and methyl ethyl ketone and the lower carboxylic acid esters as ethyl acetate. These solvents can also be used as a mixture.

As the mold releasing agents, there can be named such, for example, as stearic acid, lauric acid, and the metal salts of these acids, e.g., calcium stearate and zinc stearate. These mold releasing agents can be preferably used in an amount of 0.1 - 3% by weight based on the total of the aforesaid components (A) and (B) and, as the case may be, the component (C).

The aforementioned polymerization accelerator is effective in enhancing the molding speed or reducing the molding temperature. Preferably used as this polymerization accelerator are the organic cobalt compounds such, for example, as cobalt naphthanate and cobalt caprylate. These are preferably used in an amount of about 0.1% - about 3% by weight based on the total of the aforesaid components (A) and (B) and, as the case may be, the component (C).

On the other hand, the aforementioned polymerization retarder or stabilizer controls the molding speed and is effective in producing shaped articles with a uniform finish. For this purpose, usable are such, for example, as hydroquinone, n-propylgallate, p-benzoquinone, tetramethylthiuram disulfide and paramethoxyphenol. These are preferably used in an amount of about 0.001% - about 0.1% by weight based on the total of the aforesaid components (A) and (B) and, as the case may be, the component (C).

As the pigments, there are, for example, carbon black, ceramic black, phthalocyanin blue, phthalocyanin green, titan yellow and titan white, which pigments